Comparison of MkIV balloon and ER-2 aircraft measurements of atmospheric trace gases

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Abstract. On 8 May 1997 vertical profiles of over 30 different gases were measured remotely in solar occultation by the JPL MkIV Interferometer during a balloon flight launched from Fairbanks, Alaska. These gases included H_2O, N_2O, CH_4, CO, NO_x, NO_y, HCl, $ClNO_3$, CCl_2F_2, CCl_3F, CCl_4, $CHClF_2$, $CClF_2CCl_2F$, SF_6, CH_3Cl, and C_2H_6, all of which were also measured *in situ* by instruments on board the NASA ER-2 aircraft, which was making flights from Fairbanks during this same early May time period as part of the POLARIS experiment. A comparison of the gas volume mixing ratios in the upper troposphere and lower stratosphere reveals agreement better than 5% for most gases. The three significant exceptions to this are SF_6 and CCl_4 for which the remote measurements exceed the *in situ* observations by 15-20% at all altitudes, and H_2O for which the remote measurements are up to 30% smaller than the *in situ* observations near the hygropause.

Introduction.

During the spring and summer of 1997, a co-ordinated campaign of balloon, aircraft, and ground-based measurements of the atmospheric composition was conducted from Fairbanks, Alaska ($65^{\circ}N$, $147^{\circ}W$), in order to gain a more quantitative understanding of the reasons for ozone loss during the high latitude summer. This Photochemistry of Ozone Loss in the Arctic Region In Summer (POLARIS) campaign provided a unique opportunity to closely compare balloon profiles measured remotely by the MkIV Interferometer using solar occultation spectrometry with *in situ* profiles measured by various instruments on board the NASA ER-2 aircraft. Because the data were acquired within a few days of each other and in nearly the same location, the comparison allows the consistency of the remote and *in situ* techniques to be

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examined in an unprecedented detail.

Much of the motivation for this comparison comes from the fact that atmospheric scientists are increasingly using remote measurements (from balloon and satellite) in conjunction with aircraft *in situ* measurements to gain a more complete understanding of the atmosphere (e.g. Rex et al., this issue). Whereas the aircraft *in situ* data have a very high precision and spatial resolution, they are currently limited to altitudes below 21 km. Remotely measured profiles, despite their much poorer spatial resolution (2 km vertical, 350 km horizontal), have the virtues of extending to much higher altitudes and, in the case of satellite observations, global coverage. It is therefore essential that any biases between the two techniques be understood and documented to avoid erroneous conclusions concerning the atmospheric processes under investigation.

Previous comparisons of remote solar occultation FTIR (ATMOS) observations and *in situ* (ER-2) measurements acquired in April/May 1993 (Michelson et al. 1999) and in November 1994 (*Chang et al.*, 1996a and 1996b) showed reasonable agreement between the two techniques. Chang et al. showed that most gases (i.e. O_3, NO_y, CH_4, CCl_3F, CCl_2F_2, SF_6) agreed to better than 10%. The exceptions were H_2O (remote measurements were 10-20% smaller, particularly near the hygropause), CO (remote measurements were 25% larger in the stratosphere), CCl_4 (remote measurements were 15% larger), and HCl (remote measurements were 10-15% larger). Michelson at al. (1999) showed good (better than 10%) agreement for NO_Y, H_2O+2CH_4, O_3, HCl and CH_4. However, both of these studies were hampered by the large distances (up to 2500 km) between the ATMOS and the ER-2 measurements, and the large time separation (up to 5 weeks in Michelson et al., 1999). Both these works therefore had to rely on the constancy of tracer correlations in order to make their comparisons, and could not adequately rule out the possibility

that in some cases, atmospheric variability might have been a significant influence in the comparisons, rather than just instrumental biases.

In this work, the remote and *in situ* measurements are much more closely co-located (less than 500 km separation), reducing the effects of any atmospheric gradients, and making it more meaningful to directly compare the vertical profiles, in addition to tracer correlations. It will be shown that whereas most of the long-lived species correlate better with N_2O than altitude, for some of the shorter-lived atmospheric parameters (e.g. temperature, O_3, tropospheric H_2O, NO_X and $ClNO_3$) the opposite is true.

Further impetus for performing this comparison is provided by the expansion of measurement capabilities of both the remote and the *in situ* experiments since the 1993 and 1994 comparisons reported by *Michelson et al. (1999)* and *Chang et al.* (1996a and 1996b). CH_3Cl, $CHClF_2$, C_2H_6, $CClF_2CCl_2F$ (CFC-113), NO_x, and $ClNO_3$ have all been added to the list of gases that are compared. The latter is an important reservoir of inorganic chlorine and its *in situ* measurement from the ER-2 aircraft is the first of its kind. Also, for several of the gases reported by *Chang et al.* (e.g. H_2O, CCl_4, CCl_3F) significant improvements have been made since 1996 to the molecular spectroscopic parameters used in the analysis of the remote measurements, which are discussed in more detail later.

MkIV Experiment.

The Jet Propulsion Laboratory (JPL) MkIV Interferometer (*Toon*, 1991) is a high resolution Fourier Transform Infra-Red (FTIR) spectrometer, designed to remotely sense the atmospheric composition. Optically, it is very similar to the ATMOS instrument (Farmer, 1987) which flew four times on the Space Shuttle. The MkIV operates in solar occultation mode, in

which the sun is viewed through the Earth's limb. These limb spectra are then divided by a high-sun spectrum to remove solar and instrumental features. The resulting limb transmittance spectra are analyzed by least squares fitting, in which slant column abundances of the various gases along the limb path are determined from the depths of their absorption lines. The matrix equation relating these measured slant column abundances to the calculated slant path distances is then solved to yield the unknown volume mixing ratio (VMR) profiles. More detail on the MkIV profiles retrieval may be found in *Sen et al.* (1998).

The balloon flight analyzed in this work was launched from Fairbanks on the evening of 7 May 1997. The balloon attained float altitude of 38 km during the night, and observations were made during sunrise on 8 May 1997, which was viewed to the NE of Fairbanks. Due to the presence of a layer of cirrus cloud ahead of an approaching front, the MkIV was not able to acquire any spectra below 8 km tangent altitude. Each forward-reverse pair of MkIV spectra took 3 minutes to acquire and covered the entire 650 to 5650 cm^{-1} (1.77 to 15.4 µm) spectral region simultaneously at 0.01 cm^{-1} resolution. Therefore, all the MkIV gas profiles may be considered to be simultaneous measurements in the same airmass, unlike ATMOS which sub-divides its spectral domain between various filters.

One of the advantages of solar absorption spectrometry at high latitude is that the sunrise/set transitions take longer than at lower latitudes. On the 8 May 1997 flight, the sun took almost 2 hours to rise from a tangent altitude of 8 km to the balloon altitude of 38 km (as compared with 30 minutes at 34°N), during which time the MkIV instrument measured 39 limb spectra at an average tangent altitude separation of 0.8 km. However, the circular MkIV field of view subtends a diameter 3.6 mrad, corresponding to 1.7 km at a tangent point 480 km distant from the balloon (typical for a 20 km tangent altitude). Thus, the vertical resolution of the MkIV

profiles, estimated to be 2 km at the altitudes sampled by the ER-2, is limited primarily by the finite diameter of its field of view.

Unfortunately, the balloon flight had to be terminated immediately after the sunrise, without the possibility of a valved down descent. Therefore, only one profile was obtained from this flight. Error bars, representing the measurement precision, could nevertheless be estimated based on the quality of the spectral fits performed to determine the slant column abundances of each gas. For most gases the main systematic error arises from uncertainties in the molecular spectroscopic parameters used in the line-by-line calculations performed during the spectral fitting. The second column of Table 1 shows estimates of the size of these spectroscopic errors.

ER-2 Experiments.

Data from the following instruments were used in the comparisons discussed in this paper:

The dual-beam UV-Absorption Ozone Photometer (*Proffitt et al.*, 1989) directs radiation from a 254 nm Hg lamp through two identical sample chambers, one containing unchanged ambient air, and the other containing air scrubbed of ozone. Since ozone absorbs strongly at 254 nm, its concentration can be determined accurately (better than 5%) from the difference in the detected signals from the two chambers, together with the known 254 nm absorption cross-section.

The JPL laser hygrometer (*May*, 1998) measured the *in situ* H_2O profiles presented here. It features an open-path, multipass Herriot cell, and a tunable diode laser source operating near 1.37 μm wavelength. Measurement precision is estimated to be 0.05 ppm in the stratosphere for a 2 s integration period. Comparisons of the JPL laser hygrometer with the Harvard Lyman-alpha H_2O instrument, also on the ER-2 during POLARIS, have revealed agreement to better than 1%

above 490K (19 km) during the June/July deployment (*Hintsa*, 1999). Near the hygropause, however, the JPL instrument measured H_2O values some 5-10% larger than the Harvard instrument.

The Aircraft Laser Infrared Absorption Spectrometer (ALIAS) is a very high resolution (0.0003 cm^{-1}) scanning tunable diode laser spectrometer. By sending the laser light from four cooled, lead-salt diode lasers operating at 3.4, 6.2, 7.5, and 8.0 μm wavelength, through a 1 m long 80-pass Herriot cell, the ALIAS instrument provides fast, continuous, *in situ* measurements of N_2O, CH_4, CO, and HCl (*Webster et al.*, 1994) with total uncertainties of 3%, 5%, 10% and 10%, respectively, for typical lower stratospheric conditions.

The Airborne Chromatograph for Atmospheric Trace Species (ACATS-IV) on the ER-2 aircraft (*Elkins et al.*,1996) is designed to measure a variety of organic chlorine, bromine, and other trace species in the upper troposphere and lower stratosphere. The instrument comprises four separate gas chromatograph (GC) channels each incorporating an electron capture detector. Of the gases compared in this work, CCl_2F_2, CCl_3F, CCl_4, $CClF_2CCl_2F$, and SF_6 were measured *in situ* by ACATS, with total uncertainties of 2%, 2%, 3%, 3% and 5%, respectively, for typical lower stratospheric conditions during POLARIS (*Romashkin et al.*, manuscript in preparation). As will be discussed later, ACATS also measured N_2O and CH_4 with total uncertainties of 2% and 3% respectively.

The NOAA NO_y instrument (*Fahey et al.*, 1989) uses three independent chemi-luminescence detectors for simultaneous measurements of NO_y, NO_2, and NO. Each detector utilizes the reaction of NO in the sample with reagent O_3. The NO/O_3 reaction produces NO_2 in an excited state, which emits radiation of near 1 μm wavelength. Emitted photons are detected with a cooled photo-multiplier tube. A gold catalyst converts all forms of NO_y to NO with equal

sensitivity, prior to the detection of the NO. NO_2 is converted to NO through broadband photolysis. The total uncertainties of the NO_y and the NO_x measurements are each better than 20% for typical lower stratospheric conditions.

The Harvard University $ClNO_3$ instrument (*Bonne et al.*, 1998) thermally decomposes $ClNO_3$ into NO_2 and ClO. The latter is reacted with NO to form Cl atoms, which are then detected by resonant fluorescence. The total uncertainty of the $ClNO_3$ measurements is ±20% (1-sigma) for typical lower stratospheric conditions.

The Whole Air Sampler (WAS) consists of 29 electropolished 1.6 liter stainless steel canisters, a 4-stage metal bellows pump, a stainless steel manifold, electric valve actuators, and an electronics package for valve and pump control (*Heidt et al.*, 1989). The canisters are filled with air samples which are subsequently analyzed in the laboratory by gas chromotography. Although the WAS instrument can measure over 30 different gases simultaneously by this technique, only three, $CHClF_2$, CH_3Cl, and C_2H_6, are used in this study.

The atmospheric temperature encountered by the ER-2 aircraft was measured by the Meteorological Measurement System (MMS) with an accuracy of ±0.3K (±0.1%). This same instrument also measured the pressure to an accuracy of ±0.5 mbar.

Measurements.

During POLARIS, the NASA ER-2 aircraft carried a science payload of 16 different *in situ* instruments weighing over 2500 lbs. With a maximum operating altitude of 21 km and a possible 8 hour flight duration, the ER-2 made 21 sorties out of Fairbanks, reaching the North Pole on occasion. However, not all these data are presented in this comparison. Only ER-2 data from six sorties have been used in this study. Each sortie occurred before the break-up of the

vortex, within one week of the MkIV balloon flight, and within a few hundred km of Fairbanks (63°N<Latitude<72°N; 141°W<Longitude<157°W).

Maps of potential vorticity (e.g. Fig.1) indicate that, in early May 1997 when these measurements were made, the Arctic polar vortex was centered over Northern Europe and Russia, well away from Alaska. Although it was becoming elongated, it was still virtually intact, at least until May 12'th for altitudes below 20 km. Thus, the air in the vicinity of Fairbanks, which was sampled by MkIV and the ER-2, was extra-vortical in nature. Figure 2 show the results of trajectory analyses of airmasses arriving at the 460 K potential temperature level over Fairbanks on 8 May 1997 and points 100 km to the North, East, South, and West. Although none of these trajectories come from near the vortex, according to Figure 1, they do show quite a variety of trajectory histories, suggesting that the air over Fairbanks might nevertheless have some horizontal gradients. It will be seen later that even after binning the ER-2 data in time and location, so that vortex airmasses are excluded, there is still significant variability in the ER-2 measurements above Fairbanks, presumably due to these different airmass histories.

The situation in June/July following the mid-May break-up of the vortex, was more variable than in May. Laminae of descended remnant vortex air were observed to be entrained in the mid-latitude air near Fairbanks (*Herman et al.*, 1998). For this reason, data from a second MkIV balloon flight made from Fairbanks in early July 1997 have not been included in this comparison.

For gases which were measured by multiple instruments on board the ER-2, we have generally shown only the data from the instrument with the most data points in the region of interest. For example, the WAS instrument measures many of the gases discussed here, but only its $CHClF_2$, CH_3Cl, and C_2H_6 data are presented, because the other gases are measured more

frequently by other instruments on board the ER-2. For N_2O, which is measured by the ALIAS, ACATS, and ATLAS instruments on board the ER-2, with roughly equal quality, only the data from the ALIAS instrument are shown here. However, these three instruments displayed such excellent agreement (typically better than 2%) during the POLARIS flights used in this study, that the same conclusions would have been reached if ATLAS or ACATS data had been used instead of ALIAS data.

Figure 3a shows the locations of the ER-2 measurements (colored symbols) used in this study in relation to the MkIV measurements (black squares). Note that these remote MkIV measurement points do not represent the position of the balloon, which was slowly drifting West of Fairbanks during the flight. Instead, the black squares denote the locations of the MkIV tangent points (locations where the limb rays attained their closest approach to the Earth), which can be considered to be the effective locations of the measurements. These tangent points are plotted for integer altitudes ranging from 8 km in the far NE to 38 km in the SW, the distance between the balloon and the tangent point decreasing with increasing altitude. Black lines have been drawn through the MkIV tangent points to show the portion of the limb path that resides within 2 km vertically (and hence 175 km horizontally) of the tangent point. Typically, the limb path segments denoted by these lines will contain over 50% of the slant column abundance, and therefore the black lines illustrate the horizontal scale of the remote MkIV measurements.

In all the subsequent panels, the same colors and symbols are used to denote each data set. The ER-2 data have been plotted in reverse order of their time separation from the MkIV flight, in order to provide greater visual emphasis for the flights closest to the MkIV balloon flight. Note that there was no ER-2 flight on 8 May 1997 (970508), the day of the MkIV balloon flight, but there were flights two days before (970506) and the following day (970509).

Although Fig.3a gives the impression that the MkIV observations were fairly closely

co-located to those of the ER-2, Figure 3b shows that for the overlapping altitudes (8 to 21 km) the MkIV observations were mostly several degrees to the North of those from the ER-2. This is especially true for the later ER-2 flights (970509 and 970511). The MkIV tangent points which appear to be closest to the ER-2 points in Figure 3a were actually above 23 km altitude and are therefore not considered any further in this paper.

The MkIV temperature profile was derived by analysis of several highly temperature sensitive CO_2 lines located in the 940 cm^{-1} and 2390 cm^{-1} regions of the spectrum. Figure 3c compares this MkIV temperature profile with those measured by the MMS instrument on board the ER-2 aircraft. The MkIV profile agrees well with the ER-2 observations obtained within 2 days of the balloon flight. In particular, the temperatures measured on the 970509 ER-2 flight (orange triangles) agree extremely well (within 2°K at all levels) with the MkIV profile. Agreement with the earlier and later ER-2 flights is significantly poorer. The ER-2 profiles measured on 970430 (dark blue circles) show a considerably colder tropopause (5°K) than any of the other days. In contrast, the flight on 970511 (red triangles) shows a considerably warmer tropopause (5°K) than any of the other days. These significant temperature differences, are most likely indicative of different airmass histories, as will be confirmed later by the poor agreement of the VMRs measured on these same days. It will also be shown later that the unusual temperature maximum observed at 13.5 km altitude on 970506 and 970509 also coincided with discontinuities in the profiles of many of the long-lived tracers. This suggests that the temperature maximum marked the boundary between an airmass of sub-tropical origin which had overridden a more typical polar airmass which lay below 13.5 km altitude.

The ER-2 N_2O profiles (Figure 3d) show considerable spatial (or temporal) variability, indicating that considerably different airmasses were sampled by the ER-2 aircraft during this week of observation, despite the fact that maps of PV did not indicate the presence of strong

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horizontal gradients. The agreement between the MkIV and the ER-2 is reasonable for the two closest days, although the presence of a discontinuity at 13.5 km altitude in the ER-2 data (especially prominent on 970509) makes it difficult to assess the level of agreement in the 12 to 14 km altitude range. The poorer vertical resolution (2 km) of the remote measurements smears the discontinuity in the MkIV profile, but its presence is still discernable by the "fold" at 12 to 14 km altitude. At the upper altitudes, the MkIV profile agrees extremely well (better than 5%) with those from the ALIAS instrument, especially for the days closest to the MkIV flight. From this figure, we calculated that the average values of the ER-2/MkIV N_2O ratio was 1.03 ± 0.05. Note that the ATMOS/ER-2 intercomparison reported by *Chang et al.* (1996a and 1996b) used the *in situ* N_2O from the ATLAS instrument (Loewenstein et al. 1989), which also flew on the ER-2 during POLARIS. However, the ALIAS and ATLAS N_2O typically agreed with each other to better than 2% at all altitudes for the flights used in this study, so it would really make very little difference to the figures if ATLAS N_2O had been chosen.

Determination of Biases.

For each of the 17 species measured by both MkIV and by the ER-2, we attempted to quantify the average multiplicative biases between the *in situ* and remote measurements, and to see if they were consistent with the combined measurement uncertainties. Figure 4 illustrates the actual remote and *in situ* VMR observations that were used in this study. In the left hand panels (denoted by lower case letters) the VMR vertical profiles are plotted versus altitude, whereas in the right hand panels (denoted by upper case letters) they are plotted versus the N_2O observations described above.

These biases were initially calculated by three different methods, each of which involved

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interpolating a polynomial through the MkIV measurements so that they could be re-evaluated at any value of (i) altitude, (ii) N_2O, or (iii) Potential Temperature (PT). The bias could then be evaluated for each ER-2 measurement simply from its ratio to the MkIV polynomial evaluated at the value of an associated ER-2 independent variable (altitude, N_2O, or potential temperature). The results obtained by expressing the MkIV measurements as functions of PT were found to be almost identical to those obtained by expressing them as functions of altitude, and so for brevity the PT column has been omitted from the table. In its place, we calculated the bias as a function of N_2O^* (=$N_2O/1.03$), where the factor 1.03 is the bias of the ER-2 N_2O value with respect to MkIV, determined from the data plotted in Figure 3d. Note that although this 3% bias is well within the combined uncertainties of the ALIAS and MkIV N_2O measurements, and is apparently not statistically significant, it will be shown later that this 3% bias is in fact confirmed by the biases derived for some the other gases from their N_2O correlations.

The first column of biases in Table 1, labelled ER2/MkIV(z), are the values calculated from the data plotted in the left hand panels of Figure 4 with pressure altitude as the common independent variable. The second column of biases, labeled ER2/MkIV(N_2O), are the values calculated from the data plotted from the right hand panels of Figure 4 with N_2O as the common independent variable. The final column of Table 1, labelled ER2/MkIV(N_2O^*), are the biases that would be estimated from the right hand panels of Figure 4, if the ER-2 measurements of N_2O were first reduced by 3%, or if the MkIV measurements of N_2O were increased by 3%.

Table 1 shows that expressing the gas profiles as a function of N_2O (rather than altitude or PT) clearly reduces the standard deviation in the bias for many of the long-lived gases (e.g. CH_4, CCl_4, CCl_3F, CCl_2F_2). However, using N_2O has an undesirable side-effect: it tends to increase the bias for gases whose VMR increases with altitude (e.g. O_3, HCl, NO_x, NO_y), and to decrease the bias for gases whose VMR decreases with altitude (CH_4, CCl_2F_2, $CHClF_2$, $CHCl_3$).

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Table 1: MkIV and ER-2 measurement accuracies and calculated biases.

Gas	MkIV Accuracy	ER-2 Accuracy	Bias ER2/MkIV(z)	Bias ER2/MkIV(N_2O)	Bias ER2/MkIV(N_2O^*)	
N_2O	5%	3%	1.03±.05	(1.03±.00)	(1.00±.00)	
O_3	6%	5%	0.98±.06	1.12±.17	0.98±.14	
CO	5%	10%	0.98±.30	0.83±.35	1.11±.36	
CH_4	5%	5%	0.98±.03	0.96±.01	0.98±.01	
H_2O	6%	10%	1.25±.61	1.08±.71	1.25±.99	#
NO_y	15%	10%	1.00±.10	1.15±.09	1.03±.08	
NO_x	10%	10%	0.85±.15	0.98±.22	0.87±.21	
HCl	5%	10%	0.97±.07	1.05±.06	0.99±.06	
$ClNO_3$	15%	20%	0.71±.10	0.96±.20	0.78±.12	
CCl_4	10%	3%	0.81±.09	0.79±.07	0.85±.08	* #
CCl_3F	10%	2%	1.01±.13	0.92±.07	1.01±.08	
CCl_2FCClF_2	20%	3%	0.91±.07	0.88±.06	0.89±.05	*
CCl_2F_2	10%	2%	1.00±.06	0.96±.04	0.99±.03	
SF_6	10%	5%	0.87±.06	0.85±.06	0.86±.05	* #
$CHClF_2$	15%	5%	0.96±.03	0.91±.03	0.94±.03	*
CH_3Cl	15%	5%	1.06±.07	0.98±.06	1.05±.06	
C_2H_6	15%	5%	0.87±.21	0.85±.41	1.05±.72	

* bias differs significantly from unity.   # bias exceeds combined measurement uncertainties.

This is clearly evidence for a bias in the N_2O istself. Indeed, dividing the ER-2 N_2O values by 1.03 (final column of Table 1) yields much better agreement with the biases obtained by expressing the gas profiles as functions of altitude, yet still retains the smaller standard deviations for the long-lived gases. Thus, the biases derived from the other gases further support the adjustment of the N_2O values by the factor 1.03.

Gases whose three biases all differ from unity by more than their respective standard deviations are denoted by an asterix in Table 1. Furthermore, those gases for which the bias also exceeds the combined (RSS) measurement accuracies are denoted by a # in Table 1. Note that, for gases whose VMR increases substantially below the tropopause (e.g. H_2O, CO, C_2H_6), a large increase in the standard deviation of the bias can result from small differences in the tropopause altitude.

Remote/In-Situ Comparison.

In this section, we look in more detail at the intercomparisons illustrated in figure 4:

O_3: Figure 4a shows excellent agreement (better than 5%) for the two ER-2 flights (970506 and 970509) made closest in time to the MkIV flight, except for altitudes around 13.5 km, where the discontinuity observed in N_2O also appears in the ER-2 O_3 profiles. Interestingly, O_3, appears not to have as compact a relationship with N_2O as it does with altitude. Proffitt et al. (1992) note that binning such data points by potential temperature greatly reduces the scatter in the O_3-N_2O correlation plots, implying that in the lower stratosphere the lifetime of O_3 is long compared with transport along surfaces of constant potential temperature, but shorter than the timescales for transport across them. Also, mixing processes involving air from higher altitudes where the N_2O-

O_3 correlation is curved will introduce further scatter into the correlation. For gases with straighter correlations, these effects of mixing would be much harder to discern.

CO: Figure 4b illustrates that above 15 km the MkIV values agree well (better than 10%) with all the ER-2 flights. Below 15 km altitude the ER-2 measurements show large flight-to-flight variations, but the MkIV profile is nevertheless reasonably consistent with the two ER-2 flights (970506 and 970509) which bracket it. Plotting the CO versus N_2O (Fig.4B) reduces the scatter in the points below 15 km altitude.

CH_4: The discontinuity at 13.5 km altitude on 970509 is also apparent in the CH_4 profile (Fig.4c), and again gives rise to a fold in the MkIV profile in the 12 to 14 km altitude range. The MkIV values are larger than most of the ER-2 data, although from the vertical profiles alone this fact is not readily apparent. However, the CH_4-N_2O correlation plot (Fig.4C) clearly shows that the MkIV is biased high (or ALIAS biased low). Table 1 indicate that this is the result of a +3% bias in the ER-2 N_2O, together with a -2% bias in the the ER-2 CH_4. Interestingly, this bias seems larger for the later ER-2 flights (orange and red symbols) than the earlier flights (green and blue symbols).

H_2O: Figure 4d illustrates excellent agreement (better than 5%) above 17 km altitude and reasonable agreement in the troposphere. However, in the 12 to 16 km altitude range the MkIV measurements are at the lower bound of all the ER-2 measurements. Below the 13.5 km discontinuity, the agreement with the closest ER-2 flight (970509) is particularly poor with MkIV measuring about 3 ppm as compared with 5 ppm from the ER-2. Agreement with the second-

closest ER-2 flight (970506) is much better, but with MkIV still being about 1 ppm lower. Due to the short lifetime of tropospheric water vapor, it correlates better with altitude than N_2O below the tropopause (Fig.4d). Note that the MkIV data point at 8 km altitude and 27.8 ppm of H_2O has been clipped from the panels to allow better resolution of the stratospheric data points.

This behavior, of the remote measurements being lower than the *in situ* around the hygropause but in good agreement above and below, can also be seen in the ATMOS/ER-2 comparison reported by *Chang et al.* (1996a, Fig.3c), although it was not discussed by the authors. However, it is important to note that the NOAA Lyman-Alpha H_2O instrument (*Kelly et al.*, 1989), flown on the November 1994 ER-2 flights used in the *Chang et al.* comparison, was not flown during POLARIS. In intercomparisons with the JPL and Harvard H_2O instruments that did fly during POLARIS, the NOAA H_2O instrument consistently measured lower H_2O amounts (*Dessler et al.*, 1995; *Hintsa et al.* 1999). It therefore seems probable that if the NOAA H_2O instrument had flown during POLARIS, the agreement with the MkIV profiles would have been improved around the hygropause, but made worse above and below. Agreement with MkIV would also have been better if we had used data from the Harvard Lyman Alpha hygrometer, which measured H_2O abundances 5% to 10% lower than the JPL instrument near the hygropause (Hintsa et al., 1999).

NO_y: At the highest and the lowest altitudes, the MkIV NO_y profile agrees very well (better than 5%) with the *in situ* observations (Fig.4e). However, in the 15 to 18 km altitude range there is a tendency for the MkIV observations to lie at the lower limit of the *in situ* measurements. This bias is more noticeable in the NO_y-N_2O correlation plot (Fig.4E) which displays a 1 ppb (15%) kink at a N_2O value of 225 ppb which corresponds to 18 km altitude. Laboratory measurements

of the sensitivity of the NOAA NO_y instrument to HCN, which was measured by MkIV (0.3 ppb at 10 km; 0.2 ppb at 20 km), indicate that it is too small to be the cause of this bias, and in any case HCN has the wrong profile shape. Given the major differences in the remote and *in situ* methods of measuring NO_y (the MkIV sums individual profiles of $NO + NO_2 + HNO_3 + HNO_4 + ClNO_3 + 2xN_2O_5$, whereas the NOAA *in situ* NO_y instrument catalytically converts all the NO_y components into NO), the agreement is really very good, and well within the combined uncertainties at all altitudes.

NO_x: Although the MkIV Interferometer and the NOAA NO_y instruments both measure NO and NO_2 individually, only their sum (NO_x) is compared in Figure 4f. This is because NO and NO_2 both have substantial (>50%) diurnal variations and so a photo-chemical model would have been required to relate the remote and *in situ* NO and NO_2 due to their having been measured at different times of day. NO_x, on the other hand, has only a small diurnal variation at these altitudes (G.B.Osterman and R.J.Salawitch, private communication): less than 5% difference between sunrise, when the MkIV observations were made, and mid-morning when most of the ER-2 climb-outs occurred, and less than 15% difference between sunrise and mid-afternoon when most of the ER-2 descent profiles were measured. Therefore, a meaningful NO_x comparison can be performed without recourse to a photo-chemical model. The MkIV NO_x profile agrees very well (better than 10% at most altitudes) with the NO_x measured from the ER-2 for the two closest days. For the earliest ER-2 flight (970430), however, the NO_x values are substantially lower, probably as a result of a different airmass history.

Neither *Michelson et al.* (1999) nor *Chang et al.* (1996a) performed an ATMOS/ER-2 NO_x intercomparison because back in 1993/1994 the *in situ* NO_2 was not measured frequently

enough to provide adequate overlap with the remote measurements. This forced *Newchurch et al.* (1996) to compare ATMOS NO profiles with those measured from the ER-2 by means of a time-dependent photo-chemical model, but no comparison of NO_2 or NO_x could be performed.

HCl: Figure 4g shows good agreement (better than 10%) between the MkIV and ER-2 HCl profiles with no apparent bias. Interestingly, the *in situ* ALIAS HCl abundances are significantly lower for the earlier ER-2 flights than for the later ones. This apparent increasing trend in HCl above Fairbanks in Spring 1997 is confirmed by MkIV ground-based column observations of HCl (*Toon et al.*, 1999). Since the HCl concentration profile above Fairbanks peaks at 15 to 20 km altitude, the ground-based column abundances of HCl (the integral of the concentration profile with respect to altitude) are in fact sensitive to HCl at the same altitudes as the ER-2 measurements.

ClNO₃: Interestingly, $ClNO_3$ has a more compact correlation with altitude (Fig.4h) than with N_2O (Fig.4H), presumably through its strong dependence on O_3 (described by Sen et al., 1999), which also correlates better with altitude. Although the MkIV vertical profile of $ClNO_3$ tends to be 5% to 35% larger than those from the ER-2, this bias is less apparent from the $ClNO_3$-N_2O correlation plot. In any case, even a 35% bias would be well within the combined uncertainties of the two instruments.

CCl₄: Although both instruments measured profiles of similar shape above 11 km altitude (Fig.4i), the MkIV profile is about 15 % larger than that measured by the ER-2 on average. However, below 11 km altitude, there is a marked divergence in the CCl_4-N_2O correlation plot

(Fig.4I) which is suggestive of a problem in the MkIV data. This is likely related to the inadequacy of the molecular spectroscopic parameters in the 787-806 cm^{-1} spectral region used to measure CCl_4, where several other gases have interfering absorptions.

CCl_3F (CFC-11): The agreement, illustrated in Figure 4j, appears good (better than 10%) with little bias. Both instruments show a shallow minimum at 12 km altitude which is also seen in several of the other tracers.

$CClF_2CCl_2F$ (CFC-113): Within the fairly large MkIV uncertainties, the two sets of measurements agree at most altitudes (Fig.4k). The MkIV observations do, however, show some disturbing behavior at the lowest altitudes, which is probably related to the inadequacy of the molecular spectroscopic parameters in the 800-830 cm^{-1} spectral region used to measure $CClF_2CCl_2F$.

CCl_2F_2 (CFC-12): The agreement, illustrated in Figure 4l, is very good (better than 5%) with only a slight bias discernable from the N_2O correlation plot (Fig.4L). After correcting for the 3% bias in the ER-2 N_2O measurements, the resulting bias in CCl_2F_2 is reduced to only 1% (Table 1).

SF_6: Although the MkIV and the ER-2 profiles both exhibit a parallel decline with altitude above the tropopause (Fig.4m), there appears to be a 0.5 ppt (14%) bias with the MkIV giving the larger values. This bias is inconsistent with *Chang et al.*, (1996b) who found good agreement between ATMOS and ER-2 of SF_6 with no discernable bias. Since the ATMOS and MkIV

retrievals are based on the same laboratory cross-sections of SF_6 (*Varanasi et al.*, 1994), the different conclusions drawn by *Chang et al.* (1996a) and this study are somewhat surprising.

$CHClF_2$ (HCFC-22): The measurements, illustrated in Figure 4n, show a 0-10% bias with the remote MkIV measurements being larger. However, this bias is still within the combined uncertainties of the two measurements. Note that the ER-2 $CHClF_2$ observations presented in Figure 4n and 4N have been scaled by a factor of 1.06 in comparison with the currently archived values. This adjustment was recommended by the WAS Principal Investigator (Elliot Atlas), as a consequence of a recent re-calibration.

CH_3Cl: The agreement, illustrated in Figures 4o and 4O, is good (better than 10%) with little bias apparent. Table 1 reports a CH_3Cl bias of only 5%, after accounting for the 3% bias in N_2O.

C_2H_6: Within the previously specified latitude and date limits, ER-2 data below the tropopause are only available for one ER-2 flight (970511), when the temperature profile was quite different from the day of the MkIV profiles (970508). Nevertheless, the agreement with MkIV, illustrated in Figures 4p and 4P, is very good (better than 10%) in the troposphere, and is also reasonable in the stratosphere where the VMRs are much smaller. Note that a MkIV data point at 8 km altitude and 8.1×10^{-10} of C_2H_6 has been clipped from the figure to make it easier to see the stratospheric points.

Ozone-sonde: An Electrochemical Concentration Cell (ECC) ozone sonde was launched by NOAA/CMDL from Fairbanks later in the day of the MkIV sunrise observations (980508). This

sonde attained 33.5 km altitude, which allows comparison with the MkIV O_3 profile over a much wider altitude range than is possible with the ER-2 data. The comparison between MkIV, and the sonde, shown in Figure 5, is excellent (better than 5%) except for altitudes near the discontinuity at 13.5 km, and for a "bump" in the sonde O_3 profile from 25 to 27 km altitude. Note that the MkIV profile was measure 12 hours before the sonde profile, and that the 26 km tangent point was some 3° of latitude (300 km) to the North of Fairbanks, and so it is quite possible that the small differences in the shapes of the profiles could be real geophysical variation. No O_3-N_2O correlation plot is available for this sonde profile.

Discussion.

Much of the scatter in the *in situ* measurements at a given altitude is, in fact, real atmospheric variability, reflecting the differing origins and composition of the airmasses sampled. In fact, for some of the ER-2 flights, distinct differences are observable in the take-off and landing profiles, just 8 hours apart on the same day (e.g. CH_4 on 970506). The remote MkIV measurements, however, lack the horizontal resolution to see such non-uniformities, with the result that the MkIV VMRs are horizontally averaged, in addition to being smoothed vertically.

One of the main difficulties in directly comparing *in situ* and remote measurements is that the large differences in their spatial resolutions can reveal a significant disagreement, even when both are valid measurements. For example, the discontinuity at 13.5 km altitude in the sonde O_3 profile in Figure 5 (and in many of the ER-2 tracer profiles on 970509 and to a lesser extent on 970506) causes the sonde O_3 VMR at 13 km altitude to appear 50% larger than that measured by the MkIV, whereas at 14 km altitude the disagreement is reversed and much smaller.

To examine the true consistency of the sonde and the MkIV profiles it is necessary to

convolve the sonde profile with the MkIV averaging kernels. This was done by replacing the slant column abundances measured by MkIV with values calculated assuming that the vertical profile of atmospheric ozone everywhere along the MkIV slant paths was given by the sonde profile, and then re-retrieving the MkIV O_3 profile. The measured slant columnn uncertainties were not changed. The resulting profile, shown by the diamonds in Figure 5, is essentially what MkIV would have retrieved had the actual O_3 profile been that measured by the sonde. The close similarity between the original sonde profile and the convolved profile verifies the good (2 km) vertical resolution of the MkIV instrument. Although the smoothed sonde profile exhibits improved agreement with MkIV at 13 km altitude, it is still more abrupt than the measured MkIV profile indicating that the discontinuity at 13 km altitude was probably not as pronounced at the location of the MkIV observations as it was above Fairbanks.

For the aircraft data, is is not so simple to degrade them to the spatial resolution of the MkIV observations, because much of the variability is in the undersampled horizontal dimension rather than the vertical. However, plotting the gas profiles as functions of N_2O, rather than altitude, largely removes the effects of this mismatch in the spatial resolutions of the remote and the *in situ* measurements. This is due to the fact that the spatial resolution of the remote N_2O profile is very similar to that of the other remote gas profiles, so any discontinuities are smoothed out in a similar manner for all gases, preserving the form of the tracer correlations. This partly explains why, in Table 1, the biases of long lived tracers (e.g. CH_4, CCl_4, CCl_3F, CCl_2F_2) have a smaller standard deviation when the gas profiles are expressed as a function of N_2O rather than altitude, despite the fact that the altitude is known to a much higher accuracy and precision than the N_2O abundance.

In comparing the remote and *in situ* data, it is important to recognize the different natures

of their precision and accuracy, and the role it plays in determining which scientific problems can be addressed by the measurements. Most of the *in situ* instruments discussed here are designed to make their measurements under constant internal instrument conditions of temperature and pressure, which gives them a very high precision and an accuracy that is fairly constant with altitude. In contrast, the remote measurements have to be made under ambient external conditions which inevitably means large changes in pressure and temperature with respect to altitude. Since the shapes and strengths of most of the spectral lines used in the analysis of MkIV data are strong functions of T and P, this creates a large potential for altitude dependent error in the remote profiles. To make matters even worse, for many gases the spectral lines used at 30 km tangent altitude are far too strong to be used at 10 km altitude and so we (implicitly) switch to weaker lines. Having said this, great progress has been made in the past 10 years in reconciling laboratory spectra of pure gas samples, measured over wide ranges of temperature and pressure, with quantum mechanical calculations (*Rao and Weber*, 1992).

For gases whose VMR varies by less than a factor of two in the lower stratosphere (e.g. N_2O, CH_4), it is difficult to tell whether the observed bias is really multiplicative in nature, as assumed in the preceding analysis, or whether it is simply an offset. Errors in the spectroscopic line parameters would tend to produce a multiplicative bias in the remote measurements. Good agreement between the MkIV profiles presented here and in-situ balloon profiles measured by the ALIAS-2 instrument (Herman et al., 1998) at altitudes up to 30 km where the N_2O and CH_4 VMRs are much smaller than those presented here, suggest that a simple multiplicative scaling is appropriate. With knowledge of this correction factor, a scientist could then use both remote and *in situ* data together to test a theory or model prediction, the remote data dominating at altitudes above 21 km, and the *in situ* data below 21 km altitude. However, the H_2O bias, which

has a clear maximum at the hygropause, suggests a temperature-dependent problem in the data.

These results also indicate that for most gases, the MkIV instrument is able to make good measurements into the upper troposphere, where the data from remote limb-viewing instruments has traditionally been very poor. The temperature profile in Figure 3c indicates a tropopause altitude of 9 to 10 km for the MkIV flight on 970508, and the H_2O profiles in Figure 4d show a hygropause about 2-3 km higher. The MkIV observations would have undoubtedly gone even lower if not for the clouds. Indeed, in two separate flights from Ft. Sumner, New Mexico, under clear conditions we have obtained profiles down to 5 km altitude. However, without reliable correlative data, the accuracy of those tropospheric profiles is difficult to evaluate.

There are three main reasons why the MkIV instrument is able to do so well in the troposphere, compared with other remote instruments. Firstly the wide spectral region that it measures simultaneously (650 to 5650 cm-1) allows different absorption bands of the same gas but with widely ranging strengths to be used together. Thus, at every altitude there are absorption lines available of the optimal depth; strong lines for high altitudes, weak lines for the troposphere. This is especially important for gases whose VMR decreases with altitude (e.g. N_2O) and whose slant column abundance may fall by 3 orders of magnitude between the tropopause and 30 km altitude. Secondly, the high spectral resolution (0.01 cm-1) that can be attained by the MkIV instrument allows the pressure broadened tropospheric absorption line wings to be distinguished from the narrower stratospheric line core. This is especially important for gases (e.g. O_3, NO_2, NO, HCl) whose stratospheric absorption contribution would otherwise (i.e. at lower resolution) overwhelm the much weaker tropospheric absorption. Thirdly, the spectrometric technique allows MkIV to identify and reject regions of the spectrum where the calculated spectra do not adequately match the measured spectra, or which produce discrepant gas abundances, due perhaps to inadequacies or errors in the molecular spectroscopic database.

Conclusions.

In general, the VMR profiles retrieved by the remote solar occultation technique agree well with those measured by the various *in situ* techniques. The only significant (i.e. differences exceeding combined uncertainties) exceptions to this are SF_6 and CCl_4 for which the remote exceeds the *in situ* by 15-20% at all altitudes, and H_2O near the hygropause where the remote measurements are up to 30% smaller than the *in situ* values. Similar biases were observed in 1994 by *Chang et al.*(1996a and 1996b) for H_2O and CCl_4 in comparing ATMOS measurements with those from the ER-2. However, no bias in the SF_6 was seen in the ATMOS comparison. The ATMOS/ER-2 biases reported by *Chang et al.* (1996a and 1996b) for HCl and CO are not evident in these new data. It is noteworthy that many of the tracer correlations (e.g. CH_4, CO, H_2O, NO_y, CCl_3F, CCl_4, CCl_2FCClF_2, CCl_2F_2) are considerably improved by multiplying the MkIV N_2O values by 1.03, or by dividing the ER-2 N_2O values by 1.03. This small difference is well within the combined uncertainties of the two measurements.

For the majority of the gases considered here, their correlations with N_2O are more compact than those with altitude. This allows better determination of biases and minimizes artifacts arising from the mismatch of the vertical and horizontal resolutions of the remote and *in-situ* profiles. The exceptions to this are the shorter-lived species e.g. O_3, tropospheric H_2O, NO_x, $ClNO_3$, and temperature. A further advantage of correlating the profiles of the long-lived tracers with N_2O, rather than altitude, is that it lengthens the time separation of observations that can be meaningfully compared. For most gases, the vertical profiles plotted versus altitude display a marked deterioration in the level of agreement as the time interval between the vertical profiles being compared increased to more than 2 days. On the other hand, the tracer correlations do not deteriorate nearly as quickly, a fact that was exploited by *Michelson et al.* (1999) and by

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Chang et al. (1996a; 1996b) to perform comparisons of ATMOS and ER-2 observations seperated by large distances (2500 km) and time intervals (up to 5 weeks).

This work also demonstrates the improved viability of the solar occultation technique for space-based platforms. Although the shuttle flights of the ATMOS instrument (*Gunson et al.* 1996) showed consistently good results in the stratosphere and mesosphere, profiles retrieved at and below the tropopause displayed a high error sensitivity, especially for gases whose VMR increased with altitude. In this work we demonstrate that improvements in the hardware (dual detectors covering the entire mid-infrared simultaneously) and analysis software (multi-gas, multi-level retrieval), embodied in the MkIV experiment, extend the useful retrieval range for most gases down into the troposphere.

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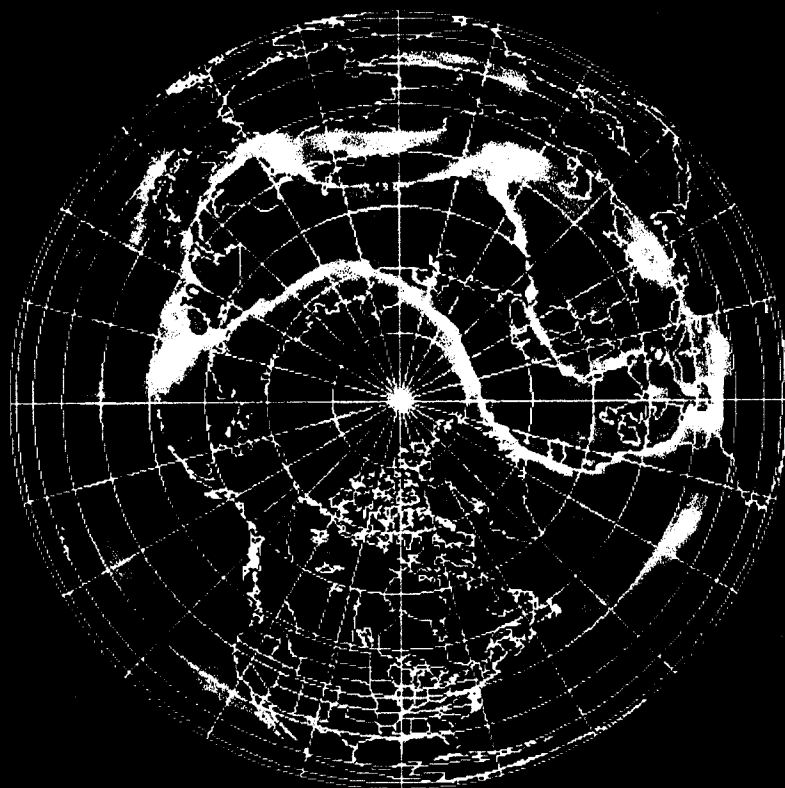
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UKMO PV at 475K

PV (10⁻⁶ m²s⁻¹ kg⁻¹)

Date: 1997/5/8 12:00

(a) Location
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(b) Latitude
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(c) Temperature
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(d) N_2O
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(M) SF₆ (ppt)
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(N) CHClF₂
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(n) CHClF₂
ER-2 970430
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ER-2 970506
MkIV 970508
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ER-2 970511

(O) CH₃Cl
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(o) CH₃Cl
ER-2 970430
ER-2 970502
ER-2 970506
MkIV 970508
ER-2 970509
ER-2 970511

(P) C₂H₆
ER-2 970430
ER-2 970502
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MkIV 970508
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